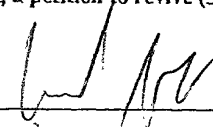


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| FORM PTO-1390<br>(REV. 12-29-99)                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   |                                                       | U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE |  | ATTORNEY'S DOCKET NUMBER<br>33126                           |
| TRANSMITTAL LETTER TO THE UNITED STATES<br>DESIGNATED/ELECTED OFFICE (DO/EO/US)<br>CONCERNING A FILING UNDER 35 U.S.C. 371                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                         |                                                       |                                                         |  | U.S. APPLICATION NO. (known, file 37 CFR 1.5)<br>09/7719116 |
| INTERNATIONAL APPLICATION NO.<br>PCT/FROO/00848                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    | INTERNATIONAL FILING DATE<br>05 April 2000 (05.04.00) | PRIORITY DATE CLAIMED<br>08/April 1999 (08.04.99)       |  |                                                             |
| TITLE OF INVENTION "PROCESS AND DEVICE FOR ANALYSIS OF RADIOACTIVE OBJECTS"                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        |                                                       |                                                         |  |                                                             |
| APPLICANT(S) FOR DO/EO/US LYOUSSI, Abdallah; PASQUALI-BARTHELEMY, Raymond;<br>PAYAN, Emmanuel & RAUX, Anne-Cecile                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  |                                                       |                                                         |  |                                                             |
| Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    |                                                       |                                                         |  |                                                             |
| <p>1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.</p> <p>2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.</p> <p>3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).</p> <p>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))</p> <p>a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</p> <p>b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau.</p> <p>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</p> <p>6. <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).</p> <p>7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))</p> <p>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</p> <p>b. <input type="checkbox"/> have been transmitted by the International Bureau.</p> <p>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</p> <p>d. <input checked="" type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</p> <p>10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</p> <p>Items 11. to 16. below concern document(s) or information included:</p> <p>11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</p> <p>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</p> <p>13. <input type="checkbox"/> A FIRST preliminary amendment.<br/><input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.</p> <p>14. <input type="checkbox"/> A substitute specification.</p> <p>15. <input type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>16. <input type="checkbox"/> Other items or information:</p> |                                                       |                                                         |  |                                                             |
| <p>"Express Mail" mailing label number <u>E1595432722US</u></p> <p>Date of Deposit <u>December 7, 2000</u></p> <p>I hereby certify that this paper or fee is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 C.F.R. § 1.10 on the date indicated above and is addressed to the Assistant Commissioner for Patents, Washington, D.C. 20231.</p> <p><u>Pamela J. Eber</u><br/>Printed Name of Person Mailing Paper or Fee</p> <p><u>Pamela J. Eber</u><br/>Signature of Person Mailing Paper or Fee</p>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |                                                       |                                                         |  |                                                             |

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|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------|----------------------------------------------|-------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------|--|
| U.S. APPLICATION NO. 097719116                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                |                     | INTERNATIONAL APPLICATION NO. PCT/FR00/00848 |             | ATTORNEY'S DOCKET NUMBER 33126                                                                                                                              |  |
| 17. <input checked="" type="checkbox"/> The following fees are submitted:<br><b>BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):</b><br>Neither international preliminary examination fee (37 CFR 1.482)<br>nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO<br>and International Search Report not prepared by the EPO or JPO ..... \$970.00<br>International preliminary examination fee (37 CFR 1.482) not paid to<br>USPTO but International Search Report prepared by the EPO or JPO ..... \$840.00<br>International preliminary examination fee (37 CFR 1.482) not paid to USPTO but<br>international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$690.00<br>International preliminary examination fee paid to USPTO (37 CFR 1.482)<br>but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... \$670.00<br>International preliminary examination fee paid to USPTO (37 CFR 1.482)<br>and all claims satisfied provisions of PCT Article 33(1)-(4) ..... \$96.00<br><b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b> |                     |                                              |             | <b>CALCULATIONS PTO USE ONLY</b>                                                                                                                            |  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |                     |                                              |             |                                                                                                                                                             |  |
| Surcharge of \$130.00 for furnishing the oath or declaration later than <input checked="" type="checkbox"/> 20 <input type="checkbox"/> 30<br>months from the earliest claimed priority date (37 CFR 1.492(e)).                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |                     |                                              |             | \$ 130.00                                                                                                                                                   |  |
| <b>CLAIMS</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | <b>NUMBER FILED</b> | <b>NUMBER EXTRA</b>                          | <b>RATE</b> |                                                                                                                                                             |  |
| Total claims                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  | - 20 =              |                                              | X \$18.00   | \$ -0-                                                                                                                                                      |  |
| Independent claims                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            | - 3 =               |                                              | X \$78.00   | \$ -0-                                                                                                                                                      |  |
| MULTIPLE DEPENDENT CLAIM(S) (if applicable)                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   |                     |                                              | + \$260.00  | \$ 270.00                                                                                                                                                   |  |
| <b>TOTAL OF ABOVE CALCULATIONS =</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                          |                     |                                              |             | \$ 1260.00                                                                                                                                                  |  |
| Reduction of 1/2 for filing by small entity, if applicable. A Small Entity Statement<br>must also be filed (Note 37 CFR 1.9, 1.27, 1.28).                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     |                     |                                              |             | \$ -0-                                                                                                                                                      |  |
| <b>SUBTOTAL =</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                             |                     |                                              |             | \$ 1260.00                                                                                                                                                  |  |
| Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30<br>months from the earliest claimed priority date (37 CFR 1.492(f)).                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     |                     |                                              |             | \$ -0-                                                                                                                                                      |  |
| <b>TOTAL NATIONAL FEE =</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   |                     |                                              |             | \$ 1260.00                                                                                                                                                  |  |
| Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be<br>accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     |                     |                                              |             | \$ -0-                                                                                                                                                      |  |
| <b>TOTAL FEES ENCLOSED =</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  |                     |                                              |             | \$ 1260.00                                                                                                                                                  |  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |                     |                                              |             | Amount to be:<br>refunded \$<br>charged \$                                                                                                                  |  |
| a. <input checked="" type="checkbox"/> A check in the amount of \$ 1260.00 to cover the above fees is enclosed.<br>b. <input type="checkbox"/> Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees.<br>A duplicate copy of this sheet is enclosed.<br>c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any<br>overpayment to Deposit Account No. 16-0820. A duplicate copy of this sheet is enclosed.<br>Our Order No. 33126                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |                     |                                              |             |                                                                                                                                                             |  |
| NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR<br>1.137(a) or (b)) must be filed and granted to restore the application to pending status.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  |                     |                                              |             |                                                                                                                                                             |  |
| SEND ALL CORRESPONDENCE TO:<br>Michael W. Garvey<br>Pearne & Gordon LLP<br>526 Superior Avenue East<br>Suite 1200<br>Cleveland, Ohio 44114<br>216-579-1700                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    |                     |                                              |             |                                                                                                                                                             |  |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |                     |                                              |             | SIGNATURE: <br>Michael W. Garvey<br>NAME: 35878<br>REGISTRATION NUMBER |  |

PROCESS AND DEVICE FOR ANALYSIS OF RADIOACTIVE OBJECTSDESCRIPTIONTechnical field

This invention relates to a process and a device for analyzing radioactive objects that use a neutronic measurement of these objects.

5 This invention can be used to analyze these objects non-destructively (in other words without affecting the physical integrity of the objects) by making active measurements (in other words controlled by external radiation) on these objects.

10 In particular, the invention is applicable to control of the radioactive product treatment process and characterization of the contents of radioactive waste packages. These packages are containers, usually made of concrete or steel, in which radioactive waste, possibly previously coated in a matrix, is placed.

15 The invention is particularly applicable to the analysis of the fissile material and/or fertile material contained in these radioactive waste packages in order to non-destructively determine the quantities of some chemical elements present in this waste.

20 It is applied directly in installations using active non-destructive analysis techniques. In particular, the analysis of the fissile material and/or the fertile material is a means of quantifying the mass of residual fuel.

25

State of prior art

Several measurement methods have been studied in order to non-destructively determine the quantity of some fissile isotopes contained in a waste package, including the neutronic interrogation technique by means of 14 MeV neutrons produced by an appropriate generator.

More particularly, measurement of prompt neutrons and delayed neutrons produced by thermal fission of the fissile material present in the waste package, is described in document US-A-4483816 (J.T. Caldwell et al).

In general, interrogation of an object by a pulsed flux of thermal neutrons is used to identify the presence of fissile material within this object. This type of method is usually used to measure fissile isotopes, namely uranium 235, plutonium 239 and plutonium 241. However, interpretation of the measurements requires prior knowledge of the isotopic composition of the fissile material.

With the technique described in the document mentioned above, the main fissile isotopes thus characterized are uranium 233, uranium 235 and plutonium 239. The various isotopes are quantified by the use of prompt and delayed signals originating from thermal neutrons. Two linear equations are then obtained. A third equation is obtained by measuring coincidences on passive neutrons (in other words neutrons emitted naturally by the material). Therefore, it is possible to calculate the various

masses of fissile isotopes mentioned above, present in the object to be measured, provided that several calibration coefficients (previously calculated) are known.

5           Nevertheless, this technique does not give any information about the presence and quantity of fertile material such as uranium 238 in the object to be analyzed.

10   Description of the invention

          The purpose of this invention is to correct this disadvantage.

          The characterization of fissile and fertile materials requires the use of an interrogating flux of  
15   thermal, epithermal and fast neutrons, since the fission threshold of uranium 238 is located at an energy of about 1 MeV. Furthermore, the contribution of uranium 238 to the measured neutronic signal can only be used for delayed neutrons emitted by fission  
20   fragments of uranium 238. Thus, the measured prompt signal corresponds to neutrons produced by thermal fission (fissile material) and the delayed signal corresponds to neutrons produced by thermal and fast fission (fissile and fertile materials).

25           This invention combines thermal, epithermal and fast interrogation with detection of prompt and delayed neutrons in order to characterize the fissile and/or fertile material that could be present in an object to be measured.

More precisely, this invention relates to a process for analyzing an object, particularly a radioactive waste package, that might contain a fissile material or a fertile material or both, the fissile material comprising M fissile isotopes and the fertile material comprising N fertile isotopes, where M and N are integer numbers equal to at least 1, this process being characterized in that:

- 10       - the object is irradiated by a neutron flux formed of thermal, epithermal and fast neutrons and resulting from a sequence of initial pulses of fast neutrons, the thermal neutrons causing fission in the fissile material and the epithermal and fast neutrons causing fission in the fissile material and in the fertile material,
- 15       - the prompt and delayed neutronic signals emitted by the object after each pulse are measured, and these signals are accumulated to obtain the sum of all signals after the last pulse,
- 20       - this sum is used to determine the contribution  $S_p$  of prompt neutrons produced by thermal fission and the contribution  $S_r$  of delayed neutrons produced by thermal, epithermal and fast fissions,
- 25       -  $S_p$  and  $S_r$  are expressed as linear combinations of the quantities, of  $M+N$  isotopes, the coefficients of these linear combinations being previously determined by calibration, and
- 30

- the quantity of each of the  $M+N$  isotopes is determined from  $S_p$  and  $S_r$  thus expressed and at least  $M+N-2$  additional items of information about quantities of  $M+N$  isotopes.

5 For example, this additional information may consist of correlations between the quantities of  $M+N$  isotopes.

According to one particular embodiment of the process according to the invention, the fissile and  
10 fertile materials contain uranium 235, uranium 238, plutonium 239 and plutonium 241.

This invention also relates to a device for analyzing an object, particularly a radioactive waste package, that may contain fissile material or fertile  
15 material or both, the fissile material containing  $M$  fissile isotopes and the fertile material containing  $N$  fertile isotopes, where  $M$  and  $N$  are integer numbers equal to at least 1, this device being characterized in that it comprises:

- 20 - means of irradiating the object by a neutron flux consisting of thermal, epithermal and fast neutrons and resulting from a sequence of initial fast neutron pulses, the thermal neutrons causing fission in the fissile  
25 material and the epithermal and fast neutrons causing fission in the fissile material and in the fertile material,
- means of counting neutrons, designed to measure prompt and delayed neutronic signals emitted by  
30 the object after each pulse, and

- means of processing the signals thus measured,  
 designed to accumulate these signals and, after  
 the last pulse, to obtain the sum of all the  
 signals, to use this sum to determine the  
 contribution  $S_p$  of prompt neutrons produced by  
 thermal fission and the contribution  $S_r$  of  
 delayed neutrons produced by thermal,  
 epithermal and fast fission, and to use  $S_p$  and  
 $S_r$  to determine the quantity of each of the M+N  
 isotopes and at least M+N-2 additional items of  
 information related to the quantities of M+N  
 isotopes, expressing  $S_p$  and  $S_r$  as linear  
 combinations of these quantities, the  
 coefficients of these linear combinations being  
 determined beforehand by calibration.

According to a preferred embodiment of the device  
 according to the invention, the irradiation means  
 comprise:

- at least one source of fast neutrons operating  
 in pulsed mode and,
- means of thermalizing these fast neutrons.

Preferably, the thermalization means comprise a  
 containment that includes a central area in which the  
 object will be placed and in which at least three sides  
 are delimited by a thickness of moderator material, the  
 neutron source being placed on a fourth side of this  
 containment and the neutron counting means being placed  
 on the three sides between the central area and the  
 thickness of moderator material, a thickness of the  
 multiplier material being provided between the central



area and the neutron source and between the central area and neutron counting means.

Each neutron counting means may also be surrounded by a thickness of neutron poison material.

5 Each neutron counting means may also be surrounded by a moderator material.

The device according to the invention may also comprise a wall made of neutron poison and moderator materials that delimits the fourth side of the  
10 containment, the thickness corresponding to the multiplier material being between this wall and the central area.

The device according to the invention may also comprise means of rotating the object within the  
15 central area of the containment.

#### Brief description of the drawings

This invention will be better understood after reading the description of example embodiments given  
20 below, which are given for guidance only and are in no way restrictive, with reference to the attached drawings in which:

- figure 1 diagrammatically illustrates the steps in a process according to the invention,
- 25 • figure 2 is a diagrammatic cutaway perspective view of a particular embodiment of the device according to the invention in an open position
- figure 3 is a diagrammatic sectional top view of the device in figure 2 in a closed position,

- figure 4 is a diagrammatic perspective sectional view of another particular embodiment of the invention, and,
- figure 5 is a diagrammatic sectional top view of the device in figure 4.

#### Detailed presentation of particular embodiments

A process according to the invention uses a thermal, epithermal and fast interrogating neutron flux in order to provoke fission reactions in an object that may contain a fissile material or a fertile material or both. This neutron flux may be obtained using at least one neutron generator operating in pulsed mode and producing fast neutrons, for example with an energy of about 14 MeV, for example using the D-T fusion reaction. An adapted thermalization cell is used to obtain a thermal, epithermal and fast neutron flux. Firstly, the thermal neutrons provoke fission reactions in the fissile material, and secondly epithermal and fast neutrons cause fission reactions in the fissile material and in the fertile material.

Furthermore, the use of a measurement method in which a signal is summated after each neutron pulse, is a means of distinguishing the contribution of prompt neutrons produced by thermal fission and the contribution of delayed neutrons produced by thermal, epithermal and fast fission, on the same signal. Only thermal fission contributes to the prompt signal since epithermal and fast fission reactions are instantaneous, therefore their contribution is drowned

in the part of the signal corresponding to interrogating neutrons.

Note that more than one pulsed neutron source can be used to increase the neutron flux and therefore the sensitivity of the measurements.

The number of fast neutron pulses may be very large and for example equal to several million. This depends on the required precision and detection limit.

The principle of a process according to the invention using a pulsed source of fast neutrons and a sequential measurement, is illustrated diagrammatically in figure 1.

Therefore, the object to be analyzed, for example a radioactive waste package, is irradiated by thermal, epithermal and fast neutrons produced by pulses from the source (and obtained as will be seen later in the description of figures 2 to 5).

Figure 1 shows the time  $t$  on the abscissa and the number of counts per second  $C(s^{-1})$  on the ordinate (on a logarithmic scale).

Neutron pulses  $I_1$  (first pulse),  $I_2$ ,  $I_3$ , ...,  $I_{n-1}$  and  $I_n$  (last pulse) are shown in the figure. The period of the generator is denoted  $T$ . The end of the last pulse occurs at an instant denoted  $T_i$ . The signal due to a single pulse denoted  $S_1$  can also be seen, together with the integrated signals due to two pulses ( $S_2$ ), three pulses ( $S_3$ ), ...  $n-1$  pulses ( $S_{n-1}$ ) and  $n$  pulses ( $S_n$ ).

Therefore, the prompt neutron signals such as  $s_p$  and delayed neutron signals such as  $s_r$  emitted after

each source pulse are measured, and these signals are accumulated. The contribution  $S_p$  of prompt neutrons produced by thermal fission and the contribution  $S_r$  of delayed neutrons produced by thermal, epithermal and fast fission, are determined from the integrated signal  $S_n$ .

Thus, two items of information  $S_p$  and  $S_r$  about the residual fuel located in the package can be determined in a single measurement.

According to the invention, these results are coupled with at least two other items of information, for example such as correlations relating the required isotope masses and obtained by calculation programs associated with operating experience in fuel reprocessing plants.

For example, it is assumed that the package contains residual uranium 235, uranium 238, plutonium 239 and plutonium 241. All this information could then be written, for example in the form of the following system of equations:

$$S_p = a_1 m(^{235}\text{U}) + a_2 m(^{238}\text{U}) + a_3 m(^{239}\text{Pu}) + a_4 m(^{241}\text{Pu})$$

$$S_r = b_1 m(^{235}\text{U}) + b_2 m(^{238}\text{U}) + b_3 m(^{239}\text{Pu}) + b_4 m(^{241}\text{Pu})$$

$$R_1 = \frac{m(^{235}\text{U})}{m(^{238}\text{U})}$$

$$R_2 = \frac{m(^{241}\text{Pu})}{m(^{239}\text{Pu})}$$

where:

$S_p$  = signal generated by prompt neutrons produced by thermal fission (count/sec.),

Sr = signal generated by delayed neutrons produced by thermal, epithermal and fast fission (count/sec.),

5  $R_1$  = correlation between the mass  $m(^{235}\text{U})$  of the uranium 235 isotope and the mass  $m(^{238}\text{U})$  of the uranium 238 isotope.

$R_2$  = correlation between the mass  $m(^{239}\text{U})$  of the plutonium 239 isotope and the mass  $m(^{241}\text{U})$  of the plutonium 241 isotope.

10  $a_i$  and  $b_i$  (where  $i$  varies from 1 to 4): calibration coefficients in  $\text{count} \cdot \text{s}^{-1} \cdot \text{g}^{-1}$ , obtained with a known object (the masses being expressed in grams).

The calibration coefficient  $a_2$  is zero since the  
15 fertile material, in the event  $^{238}\text{U}$ , does not participate in the measured signal generated by prompt neutrons.

Solution of this system gives the required masses.

The advantage of this process conform with the  
20 invention is due to the fact that the fissile material and the fertile material present in the object to be measured can be "interrogated" simultaneously making use of one or several pulsed sources of fast neutrons, for example 1 or several pulsed generators of 14 MeV  
25 neutrons.

Due to its design (examples will be given later), the device used to implement this process can produce a thermal, epithermal and fast flux while amplifying the fast component.

The contrast between the fissile material and the fertile material is thus improved.

Furthermore, the use of an associated sequential acquisition method significantly improves the sensitivity of the measurement of the delayed signal, thus overcoming the poor statistics of delayed fission neutrons. Furthermore, the combination of additional information, for example such as correlations of the different searched isotopes involving mass, molar, atomic or other ratios, is a means of separately quantifying each of the fissile and fertile isotopes present in the waste. Therefore this quantification of each isotope is obtained following a single and unique neutronic measurement on the analyzed object.

The device according to the invention as shown in the cutaway perspective view in figure 2, and in the sectional top view in figure 3, is designed to characterize an object, for example a radioactive waste container 2.

This device comprises:

- means of irradiating the container 2 by a thermal, epithermal and fast neutron flux,
- neutron counting means 4 in order to measure prompt and delayed neutron signals emitted by the container after each pulse and,
- signal processing means 6 to process the signals thus measured in order to accumulate these signals, and to use the sum of these signals to determine the contribution  $S_p$  of prompt neutrons produced by thermal fission and

the contribution  $S_r$  of delayed neutrons produced by thermal, epithermal and fast fission, and to determine the mass of each of the fissile and fertile isotopes of the waste as seen above.

The irradiation means comprise a fast neutron generator 8 operating in pulsed mode and a thermalization containment 10 for these fast neutrons in order to obtain the thermal, epithermal and fast neutron flux.

This containment comprises a central area 12 in which the container 2 will be fitted. The shape of this central area is approximately square and it is delimited by a wall 14 made of a moderator material, for example graphite.

Part 16 of this wall is mobile - for example it is installed on rails as shown in figure 2 - so that the container can be inserted in the central area.

Figure 2 shows that the containment is open whereas it is closed in figure 3 (when the container is irradiated by neutrons).

The part of the wall 14 facing the mobile part 16 comprises a space 20 in which the neutron generator 8 is housed.

The neutron count means are neutronic detection blocks 4 installed in the mobile part 16 of the wall 14 and in the two parts of the wall that are adjacent to this mobile part and are facing each other.

An element 22 made of a multiplier material, for example lead, is inserted between the generator and the

central area 12. Similarly, another element 24 made of this multiplier material is inserted between each group of detection blocks 4 and this central area.

Furthermore, each detection block 4 is surrounded by a layer 26 of neutron poison material, for example such as cadmium, and contains neutron counters, for example  $^3\text{He}$  detectors surrounded by another moderator material 28, for example polyethylene.

The containment is closed at its upper part by a graphite cover 30. It is closed at its lower part by a bottom 32 also made of graphite. This containment is also supported on a base 34, for example made of steel.

The device in figure 2 also comprises a wall 36 free to move on rails 38 fitted on base 34 so that it can be moved towards or away from the part of the wall 14 at which the generator 8 is located. This mobile wall 36 is separated from this part in the case shown in figure 2, whereas it is in contact with this part in the case shown in figure 3.

This mobile part 36 is made of neutron poison and moderator materials; for example, it may be composed of an element 40 made of graphite, coated with a boron carbide layer 42 facing the part of the wall 14 on which the generator is located.

Note that the fast neutrons emitted by the generator 8 towards the mobile wall 36 are thermalized by the graphite element 40 and are absorbed by the boron carbide layer 42 and therefore do not return to the container 2. This mobile wall 36 can be used to adjust the neutron flux.



Means of rotating this container within the central area of the containment may be provided (figure 2) in order to obtain uniform irradiation of the container 2 by neutrons. These rotation means may  
5 comprise a plate (not shown) on which the container is supported and means of rotating the plate, for example comprising a shaft 44 rigidly fixed to this plate and passing through the bottom 32 of the containment 10, and another shaft 46 rotated by a motor not shown and  
10 rotating the shaft 44 by means of gears contained in a box 48.

The block detectors 4 that are used to count the prompt signal and the delayed fission signal are preferably optimized in a known manner to optimize the  
15 sensitivity at a given energy.

Obviously, they are connected to electrical power supplies (not shown) necessary for their operation, and are also connected to signal processing means 6 located outside the containment 10.

20 The lead elements 24 that are placed in front of detection blocks 4 have a radiological shielding function. The measured containers may be very radioactive and in particular may emit high gamma radiation. It is then necessary to protect the  
25 counters so that they can be used under optimum conditions.

Neutrons output from the generator 8 enter into the lead elements 22 and 24/ and reactions of the (n, 2n) type are applied to them. This can increase the

intensity of the interrogating neutron flux by about 60%.

Each interrogating neutron can then interact in two possible ways:

5           1) The neutron is sufficiently slowed by the moderator materials, the materials in the structures and the object to be measured itself, until they reach thermal energy. It then induces fission reactions on the fissile material (for example  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Pu}$ ) in the  
10 object to be measured.

          2) The neutron is slowed but its energy is higher than about 1 MeV. It then induces fission reactions in the object to be measured, not only on the fissile material (for example  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Pu}$ ), but also on the  
15 fertile material (for example  $^{238}\text{U}$ ).

Following thermal fission, several fast neutrons (on average 2 to 3 per fission) with an average energy of 2 MeV are emitted instantaneously; these are the prompt neutrons. They are detected in blocks 4  
20 surrounded by neutron poison material, such as cadmium, that absorbs neutrons and makes them sensitive only to fast neutrons. This is a means of eliminating most of the background noise due to neutrons produced by the generator 8, that are thermal at this time of the  
25 measurement. However, the prompt neutrons signal is superposed on different background noise terms, the main terms being the "active background noise" (active signal without the contaminant) and the background noise due to passive neutron emission from the  
30 contaminant.

The measurement of prompt neutrons cannot start until the neutrons in the generator have been fully thermalized, since the signal that they induce during a few hundred microseconds after the generator pulse is very high. Consequently, all prompt neutrons produced during this first measurement phase, and particularly neutrons produced by fission reactions induced by fast neutrons from the generator, cannot be detected since they are drowned in the background noise.

The signal due to delayed fission neutrons is superposed on different background noises, the most important of which is the passive neutronic emission from the contaminant. The signal from the delayed neutrons appears to be constant during the scale of a measurement cycle, with a duration of about 10 ms, since their emission time is very long compared with this duration. They start a few hundred milliseconds to several tens of seconds after the fission reaction from which they originate following the  $\beta$ -disintegration of some fission products. Therefore, detected delayed neutrons originated from previous measurement cycles.

Delayed neutrons produced by fission reactions induced by fast neutrons contribute to the delayed neutron signal. Since the emission of a delayed neutron is delayed after the fission reaction that generated it, it is possible to detect delayed neutrons produced by fission reactions induced by fast or epithermal neutrons, or by thermal neutrons.

One important consequence is that the fertile material (for example  $^{238}\text{U}$ ) contributes to the delayed neutrons signal, but not to the prompt neutrons signal, since prompt neutrons originating from fast or epithermal fission reactions are not detectable. The effective fission cross section of this isotope at thermal energy is very small compared with the cross section of fissile isotopes, which makes its contribution to the prompt neutrons signal completely negligible since the energy spectrum of the interrogating neutrons is purely thermal during the prompt neutrons measurement.

However, the efficient fission cross section of uranium 238 is of the same order of magnitude as the fission cross section of fissile isotopes beyond 1 MeV. Furthermore, since this isotope may sometimes be present in large proportions in the contaminant, it induces a delayed signal that is not negligible compared with the signal due to fissile isotopes.

A sequential count method is used during acquisition of the signal. Thus, information originating from the contributions of fast and delayed neutrons to the total signal, for example associated with correlations such as the mass ratios of uranium isotopes 235 and 238 and plutonium isotopes 239 and 241, can be used to quantify each of the isotopes mentioned above.

Another device according to the invention is shown diagrammatically in figures 4 and 5. Figure 4

shows a perspective sectional view of this other device whereas figure 5 shows a top sectional view.

The device shown in figures 4 and 5 also includes a containment 10 comprising a central area 12 that for example will receive a radioactive waste container 2 and is delimited by four walls 50 made of a multiplier material, for example such as lead.

Neutron counters 52 are placed outside three of these walls and adjacent to these walls, and are surrounded by a moderator material, for example polyethylene. Two pulsed fast neutron generators 8 are placed outside the fourth wall 50 and adjacent to it.

As will be seen in figure 5, walls 54 made of a moderator material, for example graphite, are placed in contact with the neutron counters.

Elements 58 made of an absorbent material, for example borated polyethylene, cover the surfaces of the assembly thus obtained except for the surface on which the neutron generators are located. Furthermore, elements 60 made of a moderator material, for example polyethylene, cover the elements 58 made of an absorbent material.

Figure 5 also shows the signal processing means 6 that process signals output by neutron counters 52.

Layers (not shown) of a neutron poison material, for example cadmium, cover the neutron detectors.

A sealing layer 62, for example made of a plastic material, surrounds the walls 50.

Figure 4 shows the base 64 of the containment, which may for example be made of steel. It also shows

various thicknesses of concrete 66 surrounding the device.

Means of rotating the container may also be provided, for example comprising the rotating plate 68 that can be rotated by means of an appropriate mechanism 70, though a shaft 72 passing through the base 64.

The upper part of the device in figures 4 and 5 is covered by a steel plate 74. This plate is provided with an opening facing the central area of the containment. This opening is used to place container 2 in this area, and to take it out of the device after the measurements. Furthermore, this opening is closed by a cover 76, for example made of steel, fitted with a gripping system 78. This cover is extended downwards by an element 80 made of a moderator material, for example polyethylene.

Figure 4 also shows a fixed wall 82 made of concrete that is located facing the neutron generators 8 and that is separated from them by a space. The face of this wall 82 that is opposite the generators is fixed to a flux monitor 84 designed to determine the number of neutrons emitted by the two neutron generators 8.

Appropriate means (not shown) may be provided opposite the other face of the concrete wall 82 capable of penetrating into this device through openings (not shown), for maintenance of the device shown in figures 4 and 5.

CLAIMS

1. Process for analyzing an object (2), particularly a radioactive waste package, that might contain a fissile material or a fertile material or both, the fissile material comprising M fissile isotopes and the fertile material comprising N fertile isotopes, where M and N are integer numbers equal to at least 1, this process being characterized in that:

- 10       - the object is irradiated by a neutron flux formed of thermal, epithermal and fast neutrons and resulting from a sequence of initial pulses of fast neutrons, the thermal neutrons causing fission in the fissile material and the epithermal and fast neutrons causing fission in the fissile material and in the fertile material,
- 15       - the prompt and delayed neutronic signals emitted by the object after each pulse are measured, and these signals are accumulated to obtain the sum of all signals after the last pulse,
- 20       - this sum is used to determine the contribution  $S_p$  of prompt neutrons produced by thermal fission and the contribution  $S_r$  of delayed neutrons produced by thermal, epithermal and fast fission reactions,
- 25       -  $S_p$  and  $S_r$  are expressed as linear combinations of the quantities of  $M+N$  isotopes, the

coefficients of these linear combinations being previously determined by calibration, and

- the quantity of each of the M+N isotopes is determined from Sp and Sr thus expressed and at least M+N-2 additional items of information about quantities of M+N isotopes.

2. Process according to claim 1, in which this additional information may consist of correlations between the quantities of M+N isotopes.

3. Process according to either claim 1 or 2, in which the fissile and fertile materials contain uranium 235, uranium 238, plutonium 239 and plutonium 241.

4. Device for analyzing an object (2), particularly a radioactive waste package, that may contain fissile material or fertile material or both, the fissile material containing M fissile isotopes and the fertile material containing N fertile isotopes, where M and N are integer numbers equal to at least 1, this device being characterized in that it comprises:

- means (8, 10) of irradiating the object by a neutron flux consisting of thermal, epithermal and fast neutrons and resulting from a sequence of initial fast neutron pulses, the thermal neutrons causing fission in the fissile material and the epithermal and fast neutrons causing fission in the fissile material and in the fertile material,



- means (4, 52) of counting neutrons, designed to measure prompt and delayed neutronic signals emitted by the object after each pulse, and
- means (6) of processing the signals thus measured, designed to accumulate these signals and, after the last pulse, to obtain the sum of all signals, to use this sum to determine the contribution  $S_p$  of prompt neutrons produced by thermal fission and the contribution  $S_r$  of delayed neutrons produced by thermal, epithermal and fast fission reactions, and to use  $S_p$  and  $S_r$  to determine the quantity of each of the  $M+N$  isotopes and at least  $M+N-2$  additional items of information related to the quantities of  $M+N$  isotopes, expressing  $S_p$  and  $S_r$  as linear combinations of these quantities, the coefficients of these linear combinations being determined beforehand by calibration.

5. Device according to claim 4, in which the irradiation means comprise:

- at least one source (8) of fast neutrons operating in pulsed mode and,
- means (10) of thermalizing these fast neutrons.

6. Device according to claim 5, in which the thermalization means comprises a containment (10) that includes a central area (12) in which the object (2) will be placed and in which at least three sides are delimited by a thickness (14, 60) of moderator

material, the neutron source (8) being placed on a fourth side of this containment and the neutron counting means (4, 52) being placed on the three sides between the central area and the thickness of moderator material, a thickness of the multiplier material (22, 24, 50) being provided between the central area and the neutron source and between the central area and neutron counting means.

10           7. Device according to claim 6, in which each neutron counting means may also be surrounded by a thickness (26) of neutron poison material.

15           8. Device according to either of claims 6 and 7, in which each neutron counting means may also be surrounded by a moderator material (28).

20           9. Device according to any one of claims 6 to 8, also comprising a wall (36) made of neutron poison and moderator materials that delimits the fourth side of the containment, the thickness (22) corresponding to the multiplier material being between this wall (36) and the central area (12).

25           10. Device according to any one of claims 6 to 9, also comprising means (46, 48; 68, 70, 72) of rotating the object (2) within the central area of the containment.

ABSTRACT OF THE DISCLOSURE

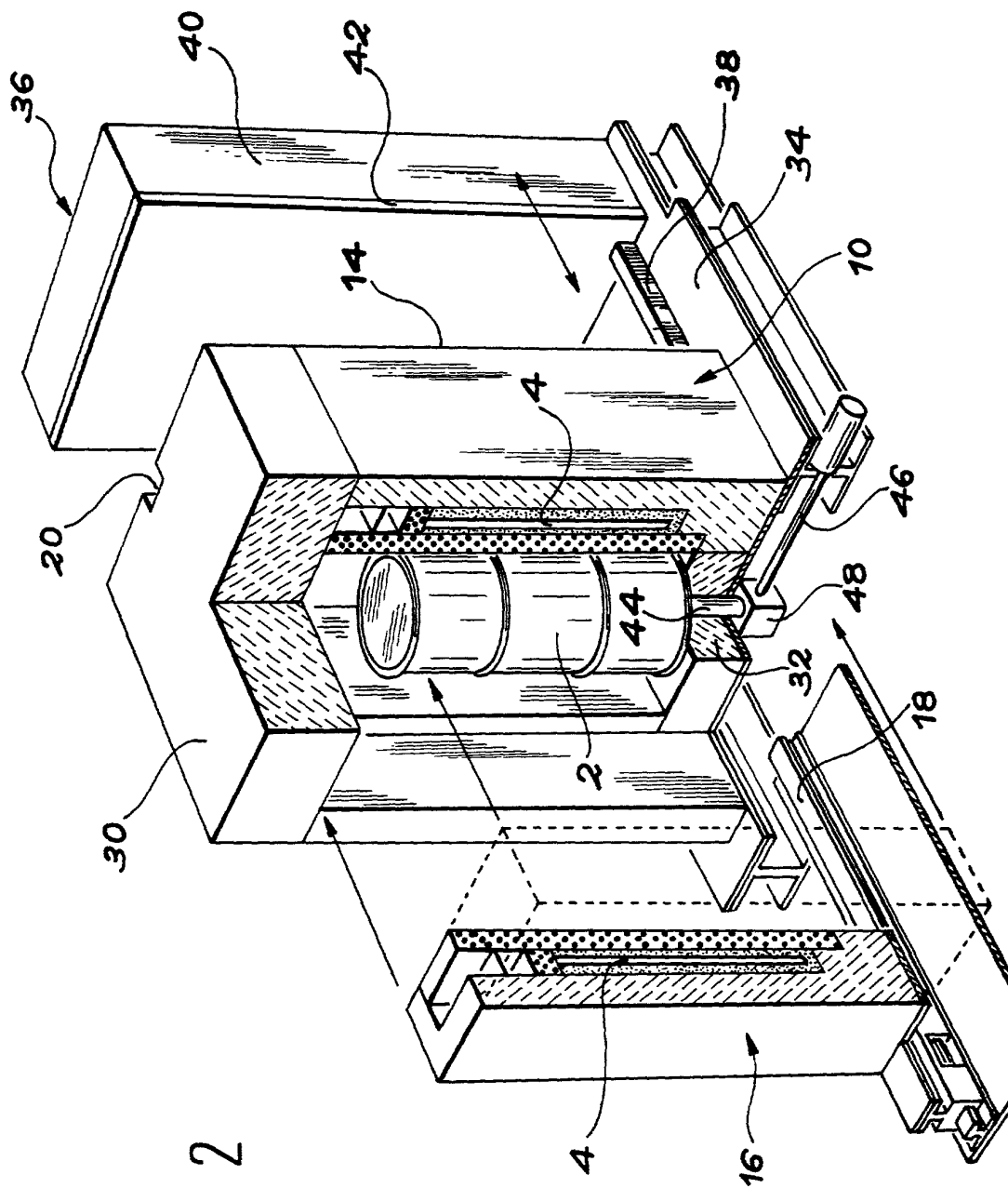
PROCESS AND DEVICE FOR ANALYSIS OF RADIOACTIVE OBJECTS

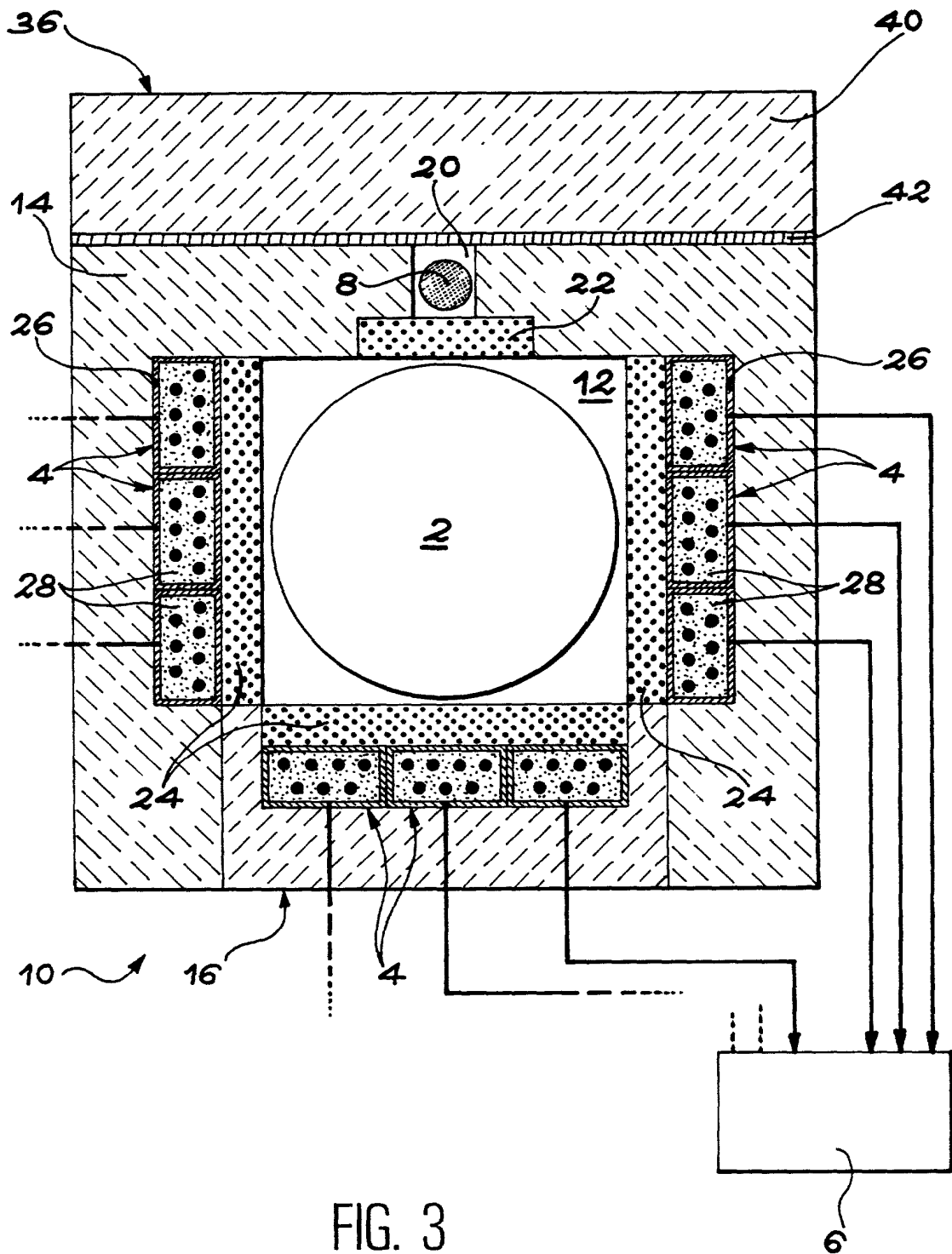
According to the invention, an object (2) and particularly a radioactive waste package that may contain fissile isotopes and/or fertile isotopes, is analyzed by irradiating the object by thermal, epithermal and fast neutrons resulting from a series of initial fast neutron pulses, the prompt and delayed neutron signals emitted by the object after each pulse are measured, these signals are accumulated, and the contribution  $S_p$  of prompt neutrons originating from thermal fission and the contribution  $S_r$  of delayed neutrons originating from thermal, epithermal and fast fission are determined from this sum of all signals, and the quantity of each isotope is determined using  $S_p$  and  $S_r$  and additional information about the isotope quantities.

Figure 2



FIG. 2





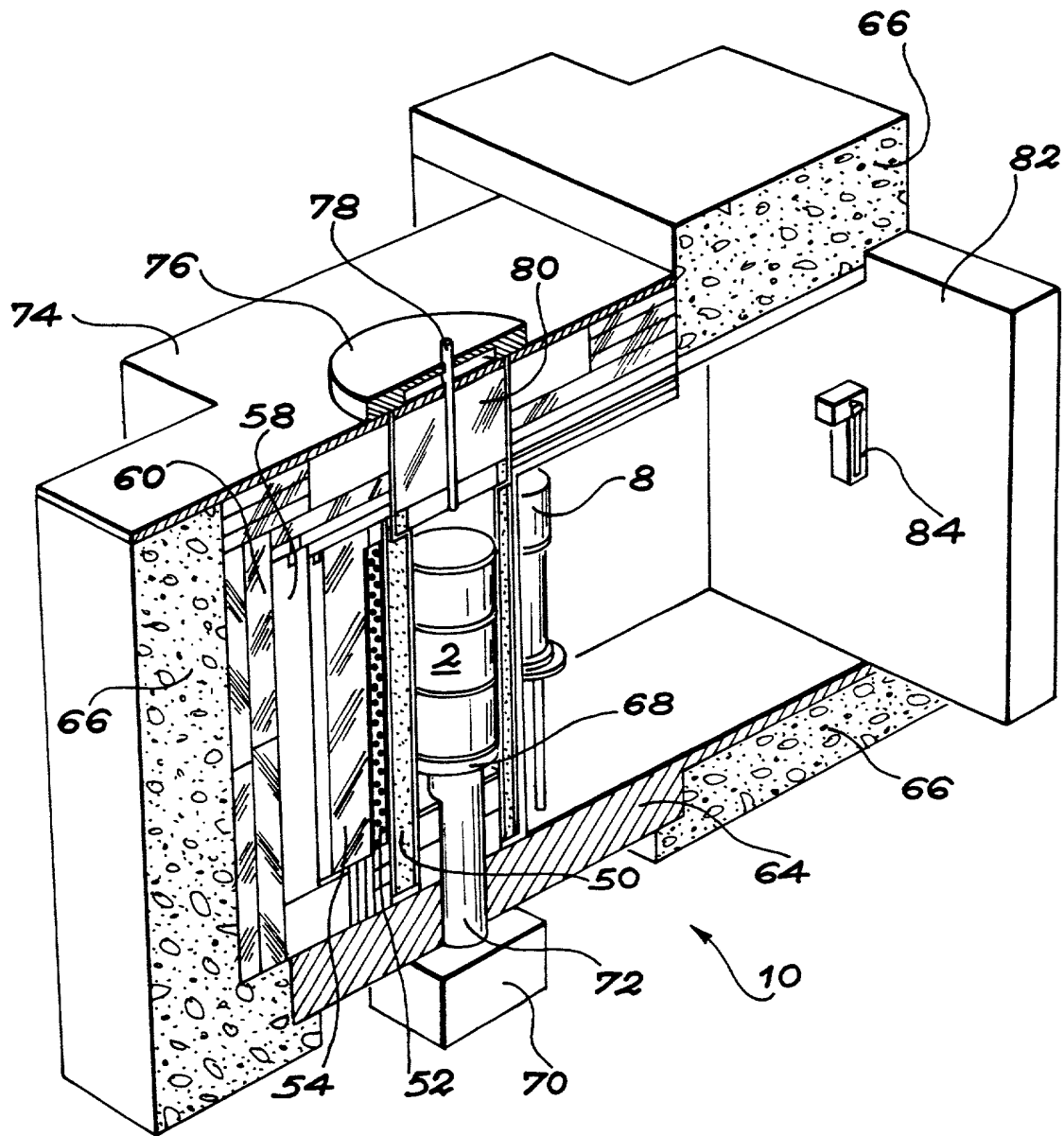


FIG. 4

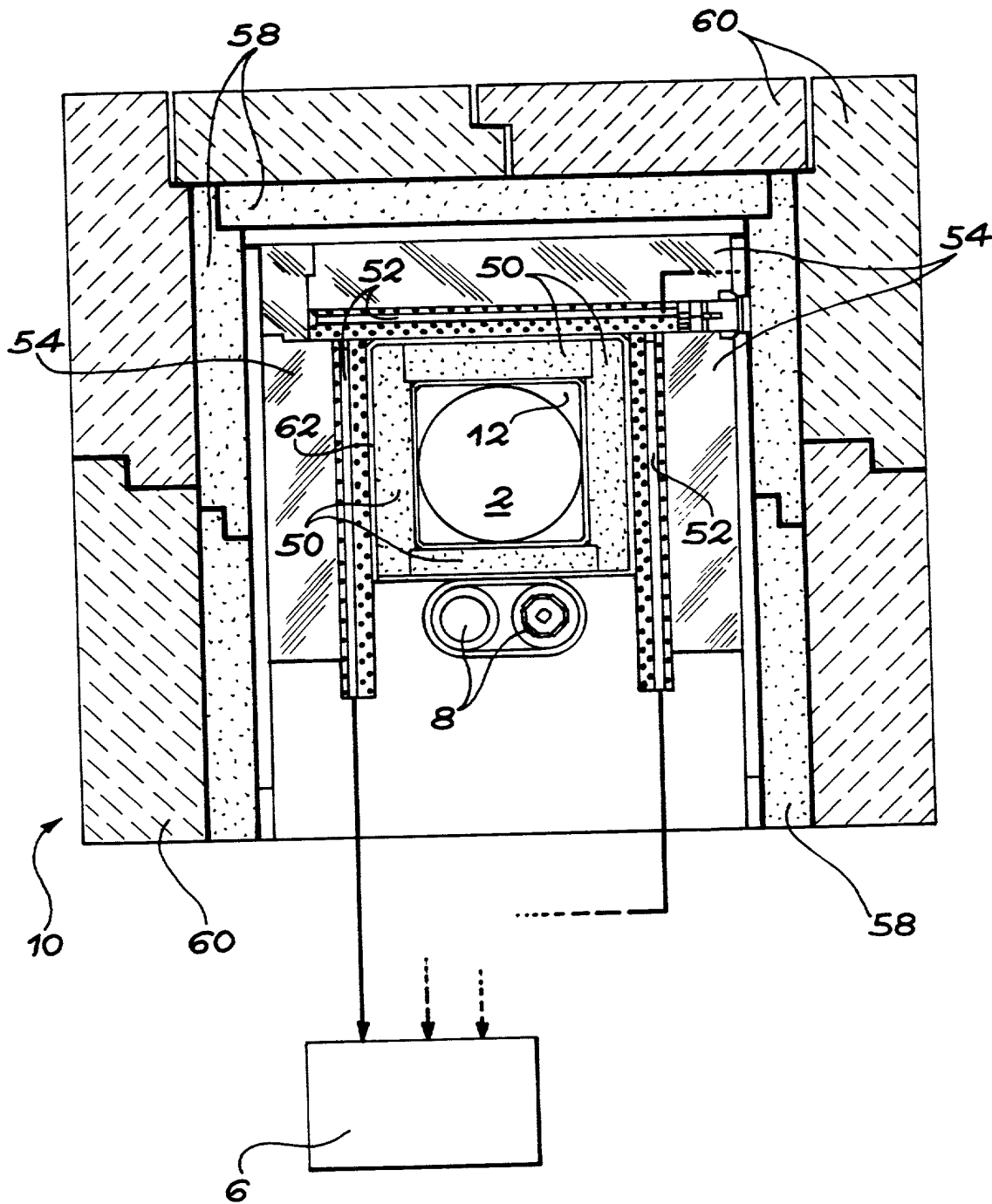


FIG. 5



# Declaration, Power Of Attorney and Petition

Page 1 of 3

WE (I) the undersigned inventor(s), hereby declare(s) that :

My residence, post office address and citizenship are as stated below next to my name,

We (I) believe that we are (I am) the original, first, and joint (sole) inventor(s) of the subject matter which is claimed and for which a patent is sought on the invention entitled

PROCESS AND DEVICE FOR ANALYSIS OF RADIOACTIVE OBJECTS.

the specification of which

☐ is attached hereto.

☐ was filed on December 07, 2000

as Application Serial No. 09/719,116

and amended on

☒ was filed as PCT international application

Number PCT/FR00/00848

on 05 avril 2000

and was amended under PCT Article 19

on

We (I) hereby state that we (I) have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We (I) acknowledge the duty to disclose information known to be material to the patentability of this application as defined in Section 1.56 of Title 37 Code of Federal Regulations.

We (I) hereby claim foreign priority benefits under 35 U.S.C. § 119 (a)-(d) or § 365 (b) of any foreign application(s) for patent or inventor's certificate, or § 365 (a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed. Prior Foreign Application (s)

| Application No. | Country | Day/month/Year | Priority Claimed                        |                             |
|-----------------|---------|----------------|-----------------------------------------|-----------------------------|
| 99 04396        | FRANCE  | 08 APRIL 1999  | <input checked="" type="checkbox"/> YES | <input type="checkbox"/> NO |
| _____           | _____   | _____          | <input type="checkbox"/> YES            | <input type="checkbox"/> NO |
| _____           | _____   | _____          | <input type="checkbox"/> YES            | <input type="checkbox"/> NO |
| _____           | _____   | _____          | <input type="checkbox"/> YES            | <input type="checkbox"/> NO |

We (I) hereby claim the benefit under Title 35, United States Code, § 119 (e) of any United States provisional application(s) listed below.

(Application Number)

(Filing Date)

(Application Number)

(Filing Date)

We (I) hereby claim the benefit under 35 U.S.C. §120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of prior application and the national or PCT International filing date of this application.

Application Serial No.

Filing Date

Status (pending, patented,  
abandoned)

And we (I) hereby appoint : Charles B. Gordon, Registration Number 16,923; William C. McCoy, Registration Number 16,885; Louis V. Granger, Registration Number 15,999; William A. Gail, Registration Number 17,409; Richard H. Dickinson Jr, Registration Number 18,622; Thomas P. Schiller, Registration Number 20,677; David B. Deioma, Registration Number 22,844; Joseph J. Corso, Registration Number 25,845; Howard G. Shimola, Registration Number 26,232; Jeffrey J. Sopko, Registration Number 27,676; John P. Murtaugh, Registration Number 34,226; James M. Moore, Registration Number 32,923; David E. Spaw, Registration Number 34,732; Michael W. Garvey, Registration Number 35,878; Paul R. Katterle, Registration Number 36,563; Richard M. Mescher, Registration Number 38,242 and Mark E. Bandy, Registration Number 35,788; our (my) attorneys, with full powers of substitution and revocation, to prosecute this application and to transact all business in the Patent Office connected therewith; and we (I) hereby request that all correspondence regarding this application be sent to the firm of PEARNE, GORDON, McCOY & GRANGER whose Post Office Address is : 526 Superior Avenue east Suite 1200 Cleveland, Ohio 44114-1484.

We (I) declare that all statements made herein of our (my) own knowledge are true and that all statements made on information and belief are believed to be true ; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such wilful false statements may jeopardise the validity of the application or any patent issuing thereon.

LYOUSSI Abdallah

NAME OF FIRST SOLE INVENTOR

Signature of Inventor

March 12, 2001  
Date

Residence : 148, Allée des Avelles

04100 - YANOSQUE.

FRANCE

Citizen of : FRANCE

Post Office Address : The same as residence

PASQUALI-BARTHELEMY Raymond

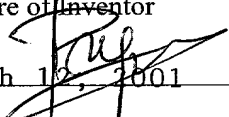
NAME OF SECOND INVENTOR

  
Signature of Inventor

March 12, 2001  
Date


PAYAN Emmanuel

NAME OF THIRD INVENTOR

  
Signature of Inventor  
March 12, 2001  
Date

RAOUX Anne-Cécile

NAME OF FOURTH INVENTOR

  
Signature of Inventor

March 12, 2001  
Date

NAME OF FIFTH INVENTOR

Signature of Inventor

Date

Residence : Rue de la Poste

05600 - Saint Crepin  
FRANCE

Citizen of : France

Post Office Address : The same as residence

Residence : Résidence "La petite

Nègresse" Bât 2, montée  
des cigales 04160 MANDOUVE

Citizen of : France

Post Office Address : The same as residence

Residence : LA SOUCHEYRE

15190, CONDAT.  
FRANCE

Citizen of : FRANCE

Post Office Address : The same as residence

Residence : \_\_\_\_\_

Citizen of : \_\_\_\_\_

Post Office Address : The same as residence